

# **EXPOSURE TO MOTOR VEHICLE EMISSIONS: AN INTAKE FRACTION APPROACH**

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## **Abstract**

Motor vehicles are a significant source of population exposure to air pollution. Focusing on California's South Coast Air Basin as a case study, I combine ambient monitoring station data with hourly time-activity patterns to determine the population intake of motor vehicle emissions during 1996 – 1999. Three microenvironments are considered wherein the exposure to motor vehicle emissions is higher than in ambient air: in and near vehicles, inside a building that is near a freeway, and inside a residence with an attached garage. Total motor vehicle emissions are taken from the EMFAC model. The 15 million people in the South Coast inhale 0.0048% of primary, nonreactive compounds emitted into the basin by motor vehicles. Intake of motor vehicle emissions is 46% higher than the average ambient concentration times the average breathing rate, because of microenvironments and because of temporal and spatial correlation among breathing rates, concentrations, and population densities.

Intake fraction (iF) summarizes the emissions-to-intake relationship as the ratio of population intake to total emissions. iF is a population level exposure metric that incorporates spatial, temporal, and interindividual variability in exposures. iFs can facilitate the calculation of population exposures by distilling complex emissions-transport-receptor relationships. I demonstrate this point by predicting the population intake of various primary gaseous emissions from motor vehicles, based on the intake fraction for benzene and carbon monoxide.

## Introduction

Motor vehicle emissions influence local, regional, and global air quality. Although strongly associated with photochemical smog and its harmful components, such as ozone and NO<sub>x</sub>, motor vehicles also contribute significantly to ambient concentrations of hazardous air pollutants and certain primary criteria pollutants, such as carbon monoxide. Nationwide, on-road motor vehicles contributed 48% of US benzene emissions in 1996 and 51% of US carbon monoxide (CO) emissions in 1999 (EPA 1999a). In the South Coast Air Basin (SoCAB) of California, on-road motor vehicles contributed 70% and 80%, respectively, of total benzene and CO emissions (CARB 2000a; SCAQMD 2000).

Previous motor vehicle exposure assessments have used measurements (e.g., ambient air monitoring stations or personal exposure monitors) and air dispersion models (e.g., the Gaussian plume or urban airshed models). While measurements and field studies are the most direct way to assess exposures, available technologies and laboratory techniques limit the number of chemicals that can be studied (Kyle *et al.* 2001; Sexton 1995). In addition, an exposure monitoring program for even a small fraction of all known hazardous air pollutants would be prohibitively expensive if applied to more than just a few individuals.

Exposure monitoring data do not directly attribute people's exposures to emission sources, yet this information is an integral part of formulating effective control strategies. Air dispersion models can overcome this limitation by tracking the concentrations, and therefore the intakes, that are attributable to each emissions source. Models enable the user to pose "what if?" questions, which can be especially useful in predicting the impact of a

specific control strategy. However, urban airshed models have only been applied to a few locations (e.g., Southern California) because of the high costs associated with gathering input data and with developing, implementing, validating, and maintaining the models. It is unlikely that sufficient resources will be available to develop detailed exposure models for motor vehicle emissions for all pollutants and locations of interest. In addition, as the complexity of exposure models increases, there is a need for metrics that enable the user to understand and interpret model results.

In this report, I present intake fractions as both a novel exposure metric and as a method for estimating exposures without relying directly on either models or measurements. I illustrate the approach by first assessing the population inhalation of benzene and CO from motor vehicles in the SoCAB (see Figure 1) during 1996 – 1999. I then apply the resulting intake fraction to other primary gaseous pollutants emitted from motor vehicles.

## Background

The intake fraction (iF) is the ratio of the total population intake of a pollutant to the total emissions (i.e., the fraction of emissions that are taken in by people). The iF depends mainly on **proximity** between the source and the receptors, **persistence** of the chemical in the environment, and the size of the exposed **population**. Persistence can refer to the time a pollutant spends in the environment in general, such as with persistent organic pollutants (POPs) that do not readily degrade. Persistence can also refer to the time a

pollutant spends in a person's breathing zone, such as with indoor pollutants in a poorly ventilated room.

Two pollutants emitted from the same source with identical fate and transport characteristics will have identical intake fractions. Likewise, two pollutants from the same *type* of source with *similar* fate and transport characteristics will have *similar* intake fractions. One would expect the iF from a given source to remain roughly constant, even as emissions change over time, as long as there are not significant net changes in the three main controlling variables (proximity, persistence, and populations).

Bennett *et al.* (2002) and Evans *et al.* (2002) review previous intake fraction research and discuss the motivation for using iF to characterize exposures. Most importantly, iFs summarize the source-to-intake relationship in a way that is easy to understand and use. iFs can be calculated using models, measurements, or both, and the concept is equally amenable to back-of-the-envelope estimates as to sophisticated analyses. Because these two papers provide recent reviews of the intake fraction literature, I do not reiterate their summaries here.

Previous investigations have emphasized the importance of motor vehicles as a dominant source of benzene and CO emissions and exposures (CARB 2000a; Duarte-Davidson *et al.* 2001; EPA 1999a; Flachsbarth 1995, 1999a; Fruin *et al.* 2001; Gonzalez-Flesca *et al.* 2000; Jo and Park 1998; Law *et al.* 1997; Macintosh *et al.* 1995; SCAQMD 2000, 2001). Macintosh *et al.* (1995) developed a probabilistic, multipathway (inhalation, ingestion, and dermal absorption) benzene exposure and dose model. For nonsmokers, they reached two main conclusions. First, population exposure to benzene is “predominantly a function of the outdoor source component of indoor air benzene levels rather than indoor

source-related exposures.” Second, uncertainty in the total dose is mainly due to uncertainty in benzene concentrations rather than to variability in time-activity patterns. Fruin *et al.* (2001) combined ambient concentration data with time-activity patterns in 14 microenvironments to assess exposure to benzene in the SoCAB. They show that the average benzene exposure for nonsmoking adults decreased from 6 ppb in 1989 to 2 ppb in 1997. They attribute this rapid decrease to comparable changes in ambient concentrations, as well as decreased exposure to environmental tobacco smoke.

To my knowledge, no published report has analyzed ambient concentration data to quantify the intake fraction. Only one previous investigation, an in press article by Evans *et al.* (2002), quantifies the intake fraction of motor vehicles. They used the Gaussian trajectory model CALPUFF, with 448 grid cells of 10,000 km<sup>2</sup> each, to calculate the iF for motor vehicle emissions on 40 highway segments throughout the United States. For primary PM<sub>10</sub>, they report values of 3-18 per million for urban locations and 1-18 per million for rural locations.

In contrast with the methodology employed by Evans *et al.* (2002), my research focuses on an urban area (17,460 km<sup>2</sup>) that would occupy less than two CALPUFF grid cells. I estimate exposures based on ambient monitoring data, rather than an air dispersion model, and I explicitly include near-source exposures. Similar to the CALPUFF methodology, I investigate exposures in areas downwind of the region wherein the emissions occur.

## Methods

For inhalation of a primary pollutant, the intake fraction can be expressed as follows:

$$\text{Intake Fraction (iF)} = \frac{\text{Total Intake}}{\text{Total Emissions}} = \frac{\int_{T_1}^{\infty} \left( \sum_{i=1}^P (C_i(t) \cdot Q_i(t)) \right) dt}{\int_{T_1}^{T_2} E(t) dt} \quad (1)$$

Here,  $T_1$  and  $T_2$  are the starting and ending times;  $P$  is the number of people in the exposed population;  $Q_i(t)$  is the breathing rate for individual  $i$  at time  $t$  ( $\text{m}^3 \text{h}^{-1}$ );  $C_i(t)$  is the incremental concentration at time  $t$  in individual  $i$ 's breathing zone that is attributable to a specific source ( $\text{g m}^{-3}$ ); and  $E(t)$  is that source's emissions at time  $t$  ( $\text{g h}^{-1}$ ). In practice, the integral in the numerator is not evaluated out to infinite times, but only until the incremental concentration attributable to the source of interest is negligibly small. For this study, it is only necessary to evaluate this integral from  $T_1$  to  $T_2$ , because the duration of that interval (four years) is several orders of magnitude longer than the time scale for pollutant transport through an urban air basin (less than a day).

iF is a dimensionless number ranging from zero, indicating that no emissions are inhaled, to one, indicating that all emissions are inhaled. An iF of one per million means that each molecule emitted to the environment has a one per million chance of being inhaled by a person. In other words, one mg of pollution is taken in (inhaled) for every kg of pollution emitted. Multiplying intake by the fraction retained in the respiratory tract yields the dose.



My method for calculating the intake fraction requires information on three parameters: emissions, population size and breathing rate, and exposure concentration. Each of these parameters is discussed below. If there were no spatial or temporal variability in the exposure concentration, the intake fraction would easily follow from equation 1. It would be computed as the product of the population size, the average breathing rate, and the average incremental exposure concentration attributable to a specific source, divided by the total emission rate for that source. However, a more detailed analysis is required for two reasons. First, publicly available concentration data comes from monitoring stations that record ambient concentrations (most monitoring stations are located on the roof of a building), rather than from exposure concentrations. Second, spatial and temporal correlations among population density, breathing rates and exposure concentrations may increase the actual population intake (Hayes and Marshall 1999).

### *Emissions*

Emissions data, which are shown in Figure 2, are based on the California's Air Resources Board's (CARB's) EMFAC database and model (CARB 2000b). I employed the 2000 version of EMFAC, which uses the latest motor vehicle emission inventory (MVEI7G) to calculate evaporative and exhaust emissions from on-road mobile sources. EMFAC databases include monthly estimates of vehicle-miles traveled and of the age distribution of the vehicle fleet. Exhaust emissions are estimated from dynamometer tests, which are run according to Federal Testing Procedure (FTP) protocols, and from CARB's database of time spent in various operating modes, such as idling, accelerating, and startup.

Evaporative emissions include drips, leaks, and “breathing losses” due to diurnal heating and cooling of the gas tank and the engine. Benzene is present in both evaporative and exhaust emissions, because it is a constituent of gasoline and also a product of incomplete combustion. Carbon monoxide is only present in exhaust emissions because it is formed during incomplete combustion but is not a gasoline constituent.

EMFAC directly estimates CO and total organic gas (TOG) emissions; it does not differentiate among the hydrocarbons that make up TOG emissions. I calculated benzene emissions by applying data from recent tunnel studies, which indicate that benzene is 3.3% of the TOG from exhaust emissions and 0.5% of the TOG from evaporative emissions (Kirchstetter *et al.* 1999a,b).

Initially there were discrepancies between the bottom-up approach of EMFAC and the top-down approach of fuel-based emissions inventories (Fujita *et al.* 1992; Harley *et al.* 1997; Pierson *et al.* 1990; Singer and Harley 1996). The former is based on scaling up the emissions from a sampling of individual motor vehicles, in terms of the emissions per km times the total km driven (Horie 1995). The latter is based on the total fuel consumption times the emissions per liter (Singer and Harley 2000). Recently versions of EMFAC agree with the fuel-based emission inventory to within about 20% (Singer and Harley 2000).

### *Population Size and Breathing Rate*

The SoCAB is home to ~15 million people occupying 6745 miles<sup>2</sup> (17,460 km<sup>2</sup>). In contrast, the population of California is ~34 million, while the population of the U.S. is ~285 million. Thus, the South Coast contains 44% of the population of California, and one

in 19 US residents. The average population density is 2,200 people mile<sup>-2</sup>, or 860 people km<sup>-2</sup>.

Using an approach based on metabolic activity (Layton 1993), the population average breathing rate has been estimated to be 12.2 m<sup>3</sup> day<sup>-1</sup>. This estimate, which incorporates information about the age distribution of the US (Census 2001), represents the average breathing rate for men, women and children. In contrast, risk assessments often use a higher breathing rate (e.g., 20 or 25 m<sup>3</sup> day<sup>-1</sup>) to allow for interindividual variability and to provide a conservative intake estimate. Figure 3 shows how the population average breathing rate was allocated to each hour of the day.

### *Exposure Concentration*

Exposure concentrations are calculated from ambient concentrations, the time spent in various microenvironments (i.e., time-activity patterns), and the concentrations in these microenvironments. Each of these three parameters is discussed below. Monthly average ambient and exposure concentrations attributable to motor vehicles are shown in Figures 4a and 4b, respectively. Figure 5 shows the typical diurnal pattern for breathing rate and exposure concentration.

*Ambient Concentrations.* The South Coast Air Quality Management District (SCAQMD) measures and records ambient pollutant concentrations at 34 air quality monitoring stations distributed throughout the South Coast Air Basin (SoCAB). During 1996 – 1999, 20 of these stations recorded one-hour average CO concentration every hour, for a total of 623,534 measurements. Six of these stations recorded 24-hour average

benzene concentration approximately twice per month, for a total of 518 measurements. Additional information on the ambient concentration data is given in Table 1. Monitoring station data and year-2000 population densities for the census tracts containing monitoring stations are combined to yield a population-weighted ambient concentration.

Because hourly ambient concentrations are available for CO but not benzene, I estimate hourly ambient benzene concentrations by applying the characteristic daily profile for CO concentrations in each month and year to the 24-hour average benzene concentration. The typical daily CO profile is shown in Figure 6. My approach assumes that benzene and CO will exhibit similar profiles over the hours of the day. This assumption is approximately true, since they are both emitted by motor vehicles, but it is not rigorously correct because CO comes from exhaust emissions while benzene comes from both exhaust and evaporative emissions. Evaporative benzene emissions will peak during hot afternoons, while CO emissions will peak during “cold start” conditions on cold mornings. The approach I have used, which does not account for these differences in the diurnal pattern between benzene and CO, is the best currently possible. If more detailed data were available on hourly ambient benzene concentrations, I would be able to refine my calculation.

One of the practical data-analysis challenges I encountered was accounting for concentrations that were below the detection limit, which occurred 5-6% of the time in the ambient CO and benzene data sets. There are several ways to utilize data with a significant fraction of nondetects. A straightforward method is to replace all non-detects with an arbitrary value, such as half the detection limit, two standard deviations below the mean, or zero. A more robust method is to replace each nondetect with a randomly generated value

below the detection limit, based on the geometric mean and geometric standard deviation of the data. I tested both methods, and, for both CO and benzene, these methods do not change the mean concentration significantly. This finding results from two conditions: the data have a small fraction of nondetects, and the detection limit is small relative to the average measured values.

*Time-Activity Patterns.* Pollutant intake depends on time-activity patterns, which indicate how much time is spent in various microenvironments. Using National Human Activity Pattern Survey (NHAPS) data provided directly to me by Klepeis (Klepeis *et al.* 2001), I examined three microenvironments: in a vehicle, in a residence with an attached garage, and all other locations, whether indoor or outdoor. In a separate analysis, described below, I also account for exposures in indoor locations that are immediately downwind of a freeway.

For the first microenvironment, I used data for the NHAPS category “in/near vehicle.” This category includes any outdoor activity that takes place inside or nearby a transportation vehicle, such as riding in a vehicle, waiting for a bus/train/automobile, and walking on a sidewalk. For the second microenvironment, I combined an estimate for the Los Angeles-Long Beach Metropolitan Area that 60% of people have an attached garage (HUD 2001) with NHAPS data on time spent in a residence. All other time was allocated to the third microenvironment, which includes both outdoor (not in/near a vehicle) and indoor (without an attached garage) locations. Of the  $1.30 \times 10^{20}$  person-hours available annually to the 15 million residents of the SoCAB, 7% is spent in-vehicle, 41% is spent inside a residence with an attached garage, and the remainder (52%) is spent elsewhere.

Other microenvironments that have been used in benzene and CO exposure assessments, such as bars or houses with natural gas cook stoves, are not needed to study exposure only to motor vehicle emissions (Fruin *et al.* 2001; Kirchstetter *et al.* 1996; Macintosh *et al.* 1995; Ott *et al.* 1992).

*Microenvironment Concentrations.* In locations that are in close proximity to motor vehicles, the exposure concentration tends to be higher than the ambient concentration. The estimated average concentration in each microenvironment is presented below. The results below are consistent with a draft EPA study that uses 37 microenvironments (EPA 2001). The increase in the exposure concentration over the ambient concentration, owing to microenvironmental enhancements, presented in Figure 5 as the normalized exposure concentration.

**In-Vehicle Concentrations.** The published literature contains many data sets of in- and near-vehicle concentrations measurements for carbon monoxide and benzene. A review of twenty two reports and journal articles on concentrations of motor vehicle pollutants inside motor vehicles indicates a high degree of variability, depending on many factors, including meteorological conditions, traffic density and speed, and emission from neighboring cars (Alm *et al.* 1999; Chan *et al.* 1991a; Chan *et al.* 1991b; Conceicao *et al.* 1997; EPA 1999b; Fernandezbremauntz and Ashmore 1995a,b; Flachsbart 1995, 1999a,b; Jo and Park 1998, 1999; Johnson 1995; Lawryk *et al.* 1995; Lioy 1992; Macintosh *et al.* 1995; McCurdy 1995; Park *et al.* 1998; Wallace 1990, 1991, 1996; Weisel *et al.* 1992). Several of these studies report both in-vehicle and ambient concentrations. Across

multiple cities and over several years of data with differing levels of ambient air pollution, typical in-vehicle CO and benzene concentrations are roughly four times greater than ambient concentrations (Flachsbart 1995, 1999b; Wallace 1996).

**Concentrations in Residences with an Attached Garage.** In an enclosed garage, evaporative emissions lead to higher concentrations of benzene but not CO. In a residence with an attached enclosed garage, these evaporative emissions can migrate into the household via airflow coupling between the garage and the living space of the house (CMHC 2001).

An enclosed garage will have slightly higher CO concentrations immediately after a vehicle pulls in or out of the garage. However, it is unlikely that there are sustained high concentrations in the garage that will enter the residential area for the following reasons. The engine is likely to be turned off soon after entering the garage, and the car will likely exit the garage soon after starting up. Short-duration, high-concentration exposures, such as the brief period a person spends inside a garage while moving from the car to the residence, do not significantly impact long term population exposures (EPA 1999b; Macintosh *et al.* 1995). In the temperate climate of Southern California, people are unlikely to “warm up” their car by running it for several minutes in the morning before driving away. Nevertheless, emissions from the car are especially high in the morning when the car is first turned on because the catalytic converter needs to warm up before it functions properly.

Unfortunately, the published literature has paid relatively little attention to this specific issue of how motor vehicle emissions in an attached garage impact

typical indoor concentrations. To adequately address this question from an experimental approach, a researcher would need to record the garage type and use level, and then measure indoor and adjacent outdoor concentrations over many days or longer. The effect of having an attached garage could show seasonal variability, due to greater emissions during “cold starts” in the winter season, so ideally the data set would span all seasons. In addition, the transport of air from the garage into the living area would likely depend strongly on the wind speed and direction. Therefore, the sampling regime would need to cover many days in order to capture a representative sample of the local meteorology. The houses would need to be selected to avoid potential sources of CO and benzene anywhere in the house, such as cigarettes or a gas stove, or in the garage, such as consumer products containing benzene or gasoline cans. Data sets that only measure indoor concentration, rather than both indoor and outdoor concentrations, are likely to be fraught with potential confounders (IES 1995). For example, houses with attached garages are likely to be more prevalent in medium density neighborhoods. In high density areas, garages are less common because of space limitations, and in low density areas, the availability of space allows for stand-alone garages. To my knowledge, no existing experimental study has addressed this question of long term, population-wide elevation of motor vehicle emissions in indoor environments owing to attached garages.

Analyzing the limited data that are available, I estimate that residences with an attached garage have vehicle-associated benzene concentrations that are ~20% higher than the ambient counterparts (Fruin *et al.* 2001; Liroy 1992; Macintosh *et al.*



1995; Thomas *et al.* 1993; Wallace 1991). On the other hand, based on no evaporative emissions and limited exhaust emissions in garages, I estimate that motor vehicle-caused CO concentrations in all houses (with or without an attached garage) are the same as the local ambient value.

**Indoor Concentrations Near Freeways.** People in houses and other buildings immediately near freeways will tend to experience higher concentrations than the ambient concentrations at monitoring stations because of the close proximity to a relatively major emissions source. I have analyzed time spent indoors near freeways separately because this microenvironment is not included in the NHAPS data. As with the previous microenvironments, the key questions are “How much higher are typical concentrations, compared to the ambient concentration?” and “How much time does the population spend in this microenvironment?” Similar to the houses with an attached garage microenvironment, using experiments to measure the impact of a freeway in a way that the results could be generalized would require careful experimental design and would likely be highly data-intensive. To my knowledge, existing data sets have not thoroughly answered this question.

Alternatively, a modeling approach can more easily estimate concentrations downwind of a generic freeway. Combining Gaussian plume model results for a line source with data showing in-vehicle concentrations as four times ambient concentrations (Fernandezbremauntz and Ashmore 1995b; Flachsbart 1995), I estimate that average concentrations of motor vehicle emissions near a major roadway are twice the basin-wide ambient concentration. This estimate accounts

for the rapid decrease in concentration immediately downwind of a major source due to atmospheric dispersion. “Near” is defined here by the distance downwind of a freeway wherein the observed concentration is significantly higher than the ambient concentration *because of that roadway’s emissions*. Drivas and Shair (1974) found this distance to be less than 100 meters. This result agrees broadly with the Gaussian plume dispersion equation for a line source (Nazaroff and Alvarez-Cohen 2001), which indicates that this distance is typically less than 300 meters. Both of these analyses assumed the wind is perpendicular to the freeway. Since all other wind directions will result in lower values for this characteristic distance, 200 meters represents a reasonable upper bound for the average characteristic distance. Combining this characteristic distance with the length of freeways in the SoCAB, 3316 km (2061 miles) (Bhat 2001) yields 660 km<sup>2</sup> of “near-freeway” land, or 4% of the total area of the SoCAB. For this portion of the analysis, I assume that the population density is uniform throughout the basin, and therefore ~4% of the people in the SoCAB are in buildings near freeways at any given time. While the assumption of uniform population density is very rough, it is sufficiently accurate for this calculation as my final results are not sensitive to this parameter. Although there are major roads that are not freeways, I have not accounted for them explicitly in this analysis because their impact on concentrations is partially reflected in the ambient concentration data.

**Concentrations in Other Locations.** In all locations other than the three microenvironments above, the exposure concentration is taken to be equal to the measured ambient concentration. Both benzene and CO are relatively nonreactive

gases, and outdoor concentrations readily penetrate into indoor environments without degradation or removal. Indoor environments may have additional sources of benzene or CO, such as gas stoves or cigarette smoke, but the existence of these sources does not alter exposure to motor vehicle emissions.

## Results

As shown in Figure 2, emissions are relatively constant throughout the year. However, concentrations of benzene and CO (Figures 4a and 4b) are about twice as high in winter as in summer. The varying concentration-to-emissions ratio generates a similar seasonal pattern in the intake fraction, as is shown in Figure 7. This variability is a consequence of varying seasonal meteorological patterns. Atmospheric transport and dispersion are lower on average during the winter because of the weaker incident solar radiation. Poorer pollutant transport means that the same emissions of primary pollutants will lead to higher ambient concentrations. Both the wind speed and the atmospheric mixing height are ~10% lower in winter (NREL 1995), leading to higher concentrations in winter. Episodes of extreme air pollution, such as the infamous "killer fog" that occurred in London during December 5 – 9, 1952, are associated with periods of highly stable meteorological conditions. As a comparison, the seasonal concentration pattern for primary pollutants – with higher concentrations in the winter – is the reverse of the pattern for ozone, which has higher concentrations in the summer due to greatly increased solar radiation.

Based on the methodology laid out above, including microenvironments, hourly ambient concentrations, breathing rates, and time-activity pattern data, I calculate annual

average intake fractions of 46 per million for CO and 49 per million for benzene. These estimates mean that approximately 50 grams of primary motor vehicle pollutants are inhaled for every million grams of primary pollutants emitted in the South Coast Air Basin. The iF for benzene is slightly higher than for CO, owing to the slightly increased exposures from attached garages, but this difference is small compared to the seasonal variability for both benzene and CO. As is shown in Figure 7, wintertime iFs are almost twice summertime iFs; this pattern is due to the seasonal variability in ambient concentrations (shown in Figure 4a). Using 48 months of data (given in Appendix 2), with a single intake fraction calculated for each month, I calculate a standard deviation of 20 per million for benzene and 15 per million for CO. Note that these standard deviations represent variations in the monthly intake fraction from the mean monthly intake fraction, not an indicator of the error associated with the estimate.

Because there are significantly more concentration data for CO than benzene, the monthly-average CO concentration measurements are more consistent from one year to the next. Consequently, my results (given in Appendix 2) show twice as much interannual variability in the iF for benzene than for CO.

## Discussion

My results are consistent with existing research. Based on previous publications, I expected the iF for an outdoor release in an urban area to be on the order of 1 – 100 per million. For example, using Gaussian plume equations, Lai *et al.* (2000) calculated an iF of 4 – 230 per million for outdoor sources, depending on the meteorology, population density, and urban area. Smith (1993) reported 20 per million as an order-of-magnitude estimate for

outdoor ground-level emission sources in urban settings. Evans *et al.* (2000) use a Gaussian plume model to calculate an intake fraction of 3.6 – 13 per million for ambient dry cleaner emissions in the US (excluding indoor exposures, such as to workers and customers). [Note that to compare the Evans *et al.* (2000) results with other values listed in this report, I have modified their reported iF of 6 – 22 per million to account for the different breathing rate they used ( $20 \text{ m}^3 \text{ day}^{-1}$  rather than  $12.2 \text{ m}^3 \text{ day}^{-1}$ ).] Schauer *et al.* (1996) reported a value of  $0.4 (\mu\text{g m}^{-3}) \text{ per } (\text{t d}^{-1})^{(i)}$  for the ratio of ambient concentration increase to emissions for elemental carbon from diesel exhaust in downtown Los Angeles. Multiplying this value by an inhalation rate of  $12.2 \text{ m}^3 \text{ day}^{-1}$  and a population of 7 million for the  $\sim 1600 \text{ km}^2$  region they used yields an iF of about 34 per million for this urban emission source. A study of Taipei City, Taiwan, stated that because of government efforts over the past two decades to relocate point sources to outside the city, over 99% of CO emissions are from motor vehicles (Chen *et al.* 2002). They present modeled and measured CO concentrations of 1.1 ppm, a population of 2.6 million people, and CO emissions of 400,000 tonnes  $\text{y}^{-1}$ . Using a breathing rate of  $12.2 \text{ m}^3 \text{ day}^{-1}$ , their results indicate an iF of 39 per million. Consistency between previous findings and the results presented here substantiates the general accuracy of my results and reinforces the potential utility of the intake fraction concept for air pollution exposure assessments.

In addition, the close agreement between the iFs for benzene and CO also reinforces the validity of the intake fraction approach. CO and benzene from motor vehicle emissions should have similar iFs because they have similar fate and transport characteristics in the atmosphere. The dominant removal mechanism from the air basin for

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<sup>(i)</sup> The units in the publication are given as  $\mu\text{g}/\text{m}^3$  per kg/day, but this is probably a typographical error.

nonreactive gases is advection, and nonreactive gases penetrate the building envelopes without impedance or removal. I characterize CO and benzene as relatively nonreactive because their characteristic lifetimes in urban atmospheres (882 hours for CO (CARB 1999) and 490 hours for benzene (EPA 1993)) are significantly greater than the typical residence time of air in the air basin (7 – 16 hours, see Appendix 1).

Within a specific air basin, the iF for any gas emitted from a broadly distributed outdoor urban source should be similar to the iF for CO and benzene if its characteristic lifetime is significantly greater than ~8 daylight hours. A gas emitted from a distributed source with a lifetime less than ~8 hours will have a smaller iF because some of the emissions will degrade before people inhale them.

For emissions with a relatively short lifetime (less than ~1 hour), a significant fraction of the total intake will occur during near-source exposures, such as the time spent in vehicles. For such a compound, it is more difficult to deduce the average concentration to which people are exposed from a limited number of ambient monitoring stations.

Further work is needed to determine the applicability of the SoCAB results to other locations. Differences in the intake fraction could arise because of differences in meteorology, such as the wind speed, rate of dispersion, and mixing height, or because of differences in the city, such as the size and population density. The intake fraction depends on the proximity between people and vehicles, which is related in a complex manner to how a city's infrastructure is set up and to social patterns that influence time-activity patterns.

### *Applying the Results for Benzene and CO to Other Chemicals*

Our understanding about exposures is built on models and measurements. Measurements are necessary to provide input data and to validate models. Properly validated models allow us to test a variety of questions and hypotheses for which experiments are difficult or impossible to undertake.

Intake fractions facilitate the application of insights gained in one investigation to other, related, investigations. For example, models and measurements can be used to calculate intake fractions initially, and then the intake fractions can then be applied to new situations or compounds beyond those specifically modeled or measured. In Table 2, I have applied my results to several chemicals that are emitted from motor vehicles. The estimation of exposure to these compounds owing to motor vehicles would be very difficult based on a pure monitoring approach. Intake fractions represent a innovative and practical method for estimating the total population intake to these chemicals due to motor vehicles.

Table 2 lists the emissions, intake fractions, and intakes for benzene plus five other compounds emitted from motor vehicles. The iF value of 0.0048%, or 48 per million, is only valid for primary pollutants that are nonreactive on the time scale of an urban air basin. Table 2 shows how intake fractions can be decreased, using the equations in Appendix 1, to account for the presence of removal reactions.

The emissions data in Table 2 are from the 1996 Toxics Emission Inventory, which uses an earlier version of EMFAC than is employed elsewhere in this report. As is discussed elsewhere in this report, earlier versions of EMFAC may underpredict emissions. It is not surprising, therefore, that benzene emissions in Table 2 are ~10% lower than the

benzene emissions reported elsewhere in this report (e.g., Table 3 and Figure 2). The ARB plans to update the inventory using the 2000 version of EMFAC; when these more current emissions estimates become available, it will be possible to update our estimates. The benzene emissions shown in Table 2 are taken from the 1996 inventory, rather than from the 2000 version of EMFAC, to maintain consistency with the data on other chemicals listed in Table 2.

### *Comparison to Ambient Concentration Analysis*

As a comparison with my main intake fraction estimate of 48 per million, I completed a second analysis using the average ambient concentration as a direct surrogate for the exposure concentration. For this simplified analysis, I computed the intake as the product of the monthly average ambient concentration, the fraction of emissions attributable to motor vehicles (70% for benzene and 80% for CO (CARB 2000a; SCAQMD 2000)), the population size, and the monthly breathing rate per person. The resulting intake fractions are  $33 \pm 14$  per million for benzene and  $32 \pm 11$  per million for CO, or 32% less than obtained by the more detailed analysis. The standard deviations presented here indicate the monthly variability in the values, rather than the confidence interval or associated level of error.

The simplified analysis, which is summarized in Table 3, agrees well with the values of 34 and 39 presented above for downtown Los Angeles and Taipei City, respectively (Chen *et al.* 2002; Schauer *et al.* 1996). Those estimates reflect a similar level of simplification in their analysis, mainly because the Schauer *et al.* (1996) and Chen *et al.* (2002) studies were investigating ambient concentrations rather than population intakes.



About half (53%) of the difference between the simplified and more detailed analysis results is due to the correlation between breathing rates and ambient concentrations, and to time spent in microenvironments. An additional 38% of the difference is due to ambient concentrations being higher in areas where the population density is higher. The remainder (9%) is due to indoor exposures near freeways.

The more detailed analysis accounts for several factors that may be important in determining the total intake, such as variability in concentration and breathing rates, and elevated concentrations in certain microenvironments. However, considerably more input data and processing time are required to complete the more complicated analysis. My study suggests that if urban population exposures for motor vehicle emissions are needed to within a error bound of approximately 50%, then the less complex analysis may be justified. Additional studies of other sources and other urban air basins are necessary to confirm this inference.

### *Exposures Occurring Outside the Air Basin*

The intake fraction results determined in this study only account for exposures that occur within the same air basin as the emissions. In reality, some exposure will occur outside of the air basin because of atmospheric transport of the pollutants from one air basin to another. I used a material balance equation based on a box model to estimate exposures occurring *outside* the SoCAB that are attributable to motor vehicle emissions occurring *inside* the SoCAB. The details of this analysis, including the equations and data used, are given in Appendix 1. I examine both a conserved pollutant and a reactive pollutant that has a characteristic lifetime of 80 hours. The characteristic lifetime for

benzene in Los Angeles is 80 hours in the summer, and 900 hours (37 days) in the winter (EPA 1993). The rate of degradation via attack by the hydroxyl radical will decrease once the air leaves the urban air basin, because of lower hydroxyl radical concentrations in less urban areas. Consequently, the lifetime of benzene will increase after leaving the SoCAB in the summer. Thus a lifetime of 80 hours, when applied to areas downwind of the South Coast, represents a compound that is more reactive than benzene in the summer.

I first look at the regional intake outside the SoCAB by assuming that motor vehicle emissions from inside the SoCAB mix throughout the two air basins that border the South Coast to the east (the Salton Sea Air Basin and the Mojave Desert Air Basin). These two air basins combined occupy 32,540 square miles, or about five times the land area of the South Coast, and they are inhabited by 1.3 million people, or less than one-tenth the population of the South Coast (CARB 2002). Next, I estimate the contribution of SoCAB motor vehicle emissions to nationwide exposure, using an area of 3,537,000 square miles and a population of 281,422,000 people (Census 2002).

For both the regional and nationwide analyses, I assume a population breathing rate of  $12.2 \text{ m}^3 \text{ person}^{-1} \text{ day}^{-1}$  and a wind speed of between  $3.36$  and  $5.50 \text{ m s}^{-1}$ . These wind speeds are the 5% and 95% values of the harmonic mean wind speed for the 75 cities listed in EPA's SCRAM database (EPA 2002). They are averaged through the mixing depth, rather than being surface wind speeds. For the regional box, I use a mixing height of between 1,200 and 12,000 meters, representing an assumption that the pollutant mixes throughout 10% to 100% of the troposphere. For the national box, I use a mixing height of 12,000 meters, representing an assumption that the pollutant mixes throughout the troposphere.

For a conserved pollutant, I calculate iFs (in units of per million) of 0.01 – 0.22 regionally and 0.20 – 0.46 nationally. For a reactive pollutant ( $k^{-1} = 80$  hours) I calculate iFs of 0.008 – 0.16 per million regionally and 0.07 – 0.08 per million nationally.

The reactive pollutant would be considered reactive when compared to the residence time of air in the US air basin (150 – 360 hours). However, it would be considered relatively nonreactive when compared to the residence time of air in the regional air basin (15 – 34 hours). This distinction leads to an important result. The regional iF for the reactive pollutant is only 20 – 40% less than the regional iF for the conserved pollutant. On the other hand, the national iF for the reactive pollutant is 3 to 5 times lower than the national iF for the conserved pollutant. *Ceritus paribus*, a more reactive compound will tend to have a smaller intake fraction.

Combining the regional and nation wide intakes, I estimate a total iF of 0.08 – 0.2 per million for a reactive pollutant and 0.2 – 0.7 per million for a nonreactive pollutant. These results for exposures *outside* the SoCAB are 70 – 620 times less than the iF for exposures *inside* the SoCAB. Consequently, for the case being studied, regional and national intakes of primary and reactive pollutants are significantly less than intraair basin intakes of urban emissions.

As a comparison, the box model is also used in Appendix 1 to predict the intake fraction for exposures occurring inside the SoCAB. The results are in the range 12 to 82 per million, which evenly brackets the values of 46 and 49 per million presented above for CO and benzene. This consistency lends support to the validity of the box model for estimating approximate intake fractions.

### *Future Work: Additional Uses for the Intake Fraction*

The iF presented in this paper reflects the total population intake. Intake fractions can also be applied to a specific individual or to the distribution of individuals' iFs within the population or a specific subpopulation. For example, intake fractions could be useful in considering environmental justice issues regarding how the total intake is distributed among the population, and whether and how the intake correlates with race or socioeconomic status.

In this report, I have investigated people's inhalation of air pollution. However, the iF could be applied to other media, exposure pathways, or endpoints. iFs could be a useful way to organize our understanding about the complex emissions-to-intake relationship for multi-media, multi-pathway compounds. For example, semivolatile organic compounds (SVOCs) are inhaled as air pollution, and they are ingested via fruits and vegetables after depositing onto crops. iF can be applied to biogenic, as well as anthropogenic, emissions. While iFs are typically thought of as applying to intakes by people, they could also be applied to other endpoints. Examples are the fraction of pollution that is taken in by a specific species of fish, or by all aquatic animals in a specific watershed. This approach could be useful in analyzing human health impacts, for example by looking at the population intake of pollutants via ingestion of seafood. It could also be useful in analyzing ecosystem or food web impacts, by looking at the fraction of emissions that are taken in by various species. Questions about ecosystem impacts often look at population dynamics, and thus a summary metric such as intake fraction could be useful in predicting or describing these impacts.

iFs are among the many metrics that can be used to compare and corroborate model predictions. As I have done in this work, iFs can be used to pose the question of whether additional levels of complication in a model or an analysis add value or modify the results. Given the increasing cost, complexity, and in some cases potential for unseen errors associated with increasingly detailed analyses, iFs can be useful in evaluating the level of detail that is appropriate for a specific situation.

### *Intake Fractions and Health Risk Assessments*

The source-by-source approach used in a conventional health risk assessment (HRA) is designed to accommodate a small number of large sources for which the local impacts are large. While it is possible to complete a conventional HRA for a situation involving many individuals and many sources, this methodology becomes more difficult as the number of sources and individuals increases. For a distributed source, such as motor vehicles in the SoCAB, whose pollution reaches many millions of people, it is important to consider the cumulative impact to the entire population. The evaluation of the health risks associated with motor vehicles represents a different context from situations typically evaluated in a health risk assessment, and the intake fraction represents a useful alternative methodology for quantifying these risks.

In order to use population intake as part of a risk assessment, it would first be necessary to convert the units for existing toxicity factors from risk per concentration to risk per intake. For example, benzene's concentration-based unit risk for leukemia is  $8.3 \times 10^{-6}$  per ( $\mu\text{g}/\text{m}^3$ ), meaning that lifetime exposure to a concentration of  $1 \mu\text{g}/\text{m}^3$  will lead to a risk of 8.3 per million (EPA 1993). This same lifetime of exposure, at an

inhalation rate of  $12.2 \text{ m}^3/\text{day}$ , will lead to a lifetime intake of 0.31 grams. Thus, the intake-based unit risk is  $27 \times 10^{-6}$  per gram, meaning that an intake of one gram will lead to a risk of 27 per million. If the dose-response curve is linear, with no threshold, then the intake-based unit risk represents the cancer risk independent of whether the intake occurs in one individual or many individuals.

Often conventional HRAs characterize environmental health risks in terms of the risk to the Maximally Exposed Individual (MEI). The MEI for a specific source is a hypothetical person who spends all of his or her time at the location of that source's maximum impact. For example, for a power plant, the MEI might be a hypothetical person who spends 100% of his or her time close to the plant and in the downwind direction. Usually the health risk to this hypothetical MEI is significantly larger than the true risk to any real individual, sometimes by orders of magnitude. If decision makers evaluate emissions sources solely in terms of the risk to a hypothetical MEI, then sources that have a large localized impact may be deemed unacceptable, whereas sources that have a moderate impact on a large number of people may be deemed acceptable. An unintended consequence of evaluating health risks based on the MEI is that a single large source that yields an unacceptable MEI risk can be deemed more acceptable if it is divided into several smaller sources, each with smaller MEI risks. The conclusion – that a larger number of smaller MEI risks is acceptable – may be reached *even though the total risk to the exposed population could remain unchanged or even increase* through the process of splitting a large source into many smaller ones.

A second method commonly used in a conventional HRA is to identify the number of people above a certain concentration or risk level. This method is more useful than the

MEI approach at capturing the population exposure, but it is still not complete. Two sources could have the same number of people above a certain threshold, while exposing the above-threshold and the below-threshold populations to very different concentrations. In addition, similar to the MEI approach, a single source split up into many little sources may cease to expose people above a certain level, even if doing so does not decrease the total population health burden.

In contrast to a conventional HRA, intake fractions are used to calculate the total population intake, which (for some pollutants) is more closely associated with the cumulative risk to the population. For a compound exhibiting a linear, no threshold, dose-response relationship, the population's health risk is directly proportional to the total population intake. The use of intake fractions in risk assessments shifts the framework from one based on the risk to an individual or group of individuals to one based on the risk burden to an entire population or to a subpopulation.

Intake fractions summarize the emissions-to-intake relationship in a way that is easy to understand and easy to use. One could imagine a catalogue of iFs, organized by pollutant type (primary nonreactive, primary reactive, or secondary) and by source type, which a risk assessor could draw on for various situations.

Finally, intake fractions represent a novel way of quantifying environmental health questions, and they can be useful for comparing pollutant impacts across diverse source categories. For example, using total population intake as a metric facilitates comparing motor vehicles to tobacco smoke as sources of exposure to benzene.

## Uncertainty and Sensitivity Analysis

My research quantifies the population intake of motor vehicle emissions, a parameter that not possible to measure directly. An uncertainty analysis is important to determine the confidence that can be placed in the results and methodology. In addition to the uncertainty analysis presented here, the uncertainty in the results could also be ascertained by comparing these results with future research that uses an alternative method to quantify the population intake of motor vehicle emissions.

Systematic errors can reduce the accuracy of a measurement and random errors can reduce the precision of a measurement. Systematic errors refer to biases that lead to consistent under- or over-estimation, while random errors are fluctuations which lead to a specific measurement being too high or too low. For example, if a laboratory technique to measure the concentration of a specific compound in an air sample has random errors but no systematic errors, then duplicate measurements of a single air sample will yield different results while the average of many duplicate samples is likely to yield the correct answer. This technique is accurate but not precise, because the average is correct even though any individual measurement is not necessarily correct. In contrast, if a laboratory technique has systematic errors but no random errors, then duplicate measurements of a single sample will yield similar results, all having a similar level of error. This technique is precise but not accurate, because even though there is a high level of consistency between measurements the average of several measurements is different from the correct value.

Among the data that I use in this paper, four parameters dominate the results. These four parameters are emissions from motor vehicles, ambient concentrations, the size of the population, and the breathing rate. In addition, a second set of parameters is relatively



important, though less so than the previous four. This second list is concentrations in vehicles, time spent in vehicles, concentrations inside residences with an attached garage, and population density at each of the ambient monitoring stations.

Below, I first complete an uncertainty analysis for the four input parameters that dominate my results (emissions, ambient concentrations, population size, and breathing rate). Then, I conduct a crude sensitivity analysis for the four additional parameters (concentrations in vehicles, time in vehicles, concentrations in residences with an attached garage, and population density at each monitoring station). Finally, I provide a qualitative discussion of methodological uncertainties.

### *Uncertainty Analysis*

*Emissions.* The EMFAC emissions model, which yields monthly emissions estimates, is maintained by the California Air Resources Board (CARB). CARB does not provide an uncertainty estimate for the EMFAC results. A comparison between EMFAC and a fuel-based emission inventory (Singer and Harley 2000) indicates agreement to ~20%, with the former methodology yielding lower estimates. This level of agreement is a significant improvement from previous versions of EMFAC, which were a factor of 2 – 4 lower than fuel-based emission inventories (Fujita *et al.* 1992; Harley *et al.* 1997; Pierson *et al.* 1990; Singer and Harley 1996).

*Ambient concentrations.* Because of the relatively large number of measurements (623,534 CO measurements and 518 benzene measurements), random error in individual measurements will tend to cancel out and not bias the final result. However, the potential exists for systematic error in the measurement technique.

The CO and benzene monitoring stations are audited on a regular basis using a laboratory-prepared calibration sample of a known concentration. During the years considered (1996 – 1999), audits of monitors throughout California yielded an average percent difference between the calibration sample and the monitor's measurement of 0.5% and -11% for CO and benzene, respectively (CARB 2001; Miguel 2002). These audits indicate that CO monitors have a high degree of accuracy while benzene monitors tend to underestimate the true concentration somewhat.

*Population size.* The US Census Bureau does not directly report uncertainty. An indirect indicator of the level of accuracy in their data is the undercount rate, which is the amount by which survey tally results are increased to account for people not surveyed. Publicly available census data have been corrected to account for the undercount rate. In California and the US, the undercount rates are 2.7% and 1.6%, respectively. In Los Angeles, Orange, Riverside, and San Bernardino Counties, the rates are 3.3%, 2.1%, 2.4%, and 2.6%. These values indicate a high degree of precision and accuracy in census results.

*Breathing rate.* There is no direct information on the accuracy and precision of the breathing rate data I use (Layton 1993). Variability in breathing rates offers a proxy for the level of accuracy. Lifetime average rates for men and women are  $14.1$  and  $10.2 \text{ m}^3 \text{ d}^{-1}$ , respectively. These two values are  $2.0 \text{ m}^3 \text{ d}^{-1}$  different from the average breathing rate of  $12.2 \text{ m}^3 \text{ d}^{-1}$ . Breathing rates for people 18 and under are  $11.2 \text{ m}^3 \text{ d}^{-1}$  while breathing rates for people over 18 are  $12.5 \text{ m}^3 \text{ d}^{-1}$ . These two values are  $1.0$  and  $0.4 \text{ m}^3 \text{ d}^{-1}$  different from the average breathing rate. Based on these relatively narrow ranges of breathing rates in Layton's (1993) results, I conclude that the accuracy in my population-weighted breathing rate is  $12.2 \pm 1 \text{ m}^3 \text{ d}^{-1}$  or better. This level of accuracy represents an error of 8% or less.

Combining the uncertainties from the above four values, if the breathing rate is too high by 8%, the census values are too high by 3.3%, the concentrations are correct, and the emission inventory is too low by 20%, then the actual iF will be 26% lower than my results. Conversely, if the breathing rate is too low by 8%, the census values are too low by 3.3%, the benzene concentrations are too low by 11%, and the emission inventory is correct, then the correct benzene result will be 24% higher than my result. If the breathing rate is too low by 8%, the census values are too low by 3.3%, the CO concentrations are correct, and the emission inventory is correct, then the correct CO result will be 12% higher than my result. These values provide a bounding estimate on the magnitude of error likely to be associated with my result. They are likely to be hard bounds because the errors are unlikely to line up at maximum possible amounts. Based on this analysis, I conclude that my results are accurate to  $\pm 25\%$  or better, and that the CO results are somewhat more certain than the benzene results because of greater accuracy in the ambient concentration data.

### *Sensitivity Analysis*

The above estimate of 25% uncertainty includes the four factors that most strongly influence the results. However, it does not include the uncertainty in several inputs, such as time-activity pattern data and concentrations in microenvironments, because the level of uncertainty associated with these data has not been quantified. Among the data for which uncertainty has not been quantified above, the most important values are the concentration and time spent in vehicles, the concentration inside residences with an attached garage, and the population density around the air basin throughout the day. A crude sensitivity analysis

indicates the potential importance of uncertainty in these variables. The results of this sensitivity analysis are shown in Table 4. For example, changing the in-vehicle concentration by 20% yields a 6% change in the intake fraction results. Doubling the population density at each specific monitoring station changes the intake fraction results by between -2% and 8%. Note that one station (Station #60371301, located at 11220 Long Beach Boulevard in Lynwood, California) has both the highest population density and the highest annual average concentration. Changes to the population density for this station have a larger impact on the intake fraction than changes to other stations.

### *Methodological Uncertainties*

The above estimate of 25% uncertainty also does not include the potential for methodological errors, which can be especially difficult to quantify in a rigorous manner. For example, I use census data on population density to population-weight the ambient concentration measurements. These data account for where people live, but not where they travel during the day (i.e., downtown to shop or work). Given the enormous task (and uncertainty) associated with attempting to estimate population densities as a function of time, this omission seems appropriate. A second example of a potential methodological error in my work is how I implemented the NHAPS data on time spent in vehicles. Specific data on time spent “in an automobile” rather than simply “in a vehicle” (which includes trains and airplanes) is not currently available in NHAPS. By including time spent in trains and airplanes with time spent in a motor vehicle, I will slightly overpredict the average exposure to motor vehicle exhaust. However, the magnitude of this error is likely to be small, and unlikely to significantly impact my results. A third example of a potential

methodological error is the use of monitoring stations to capture ambient concentrations of benzene and CO. Monitoring stations offer the most comprehensive ambient concentration data set available. Nevertheless, the methodology used in this report, which estimates exposure concentrations based on monitoring station data, may under- or over-estimate exposures. For example, the methodology will over-estimate exposures if monitoring station locations are, on average, closer to roadways than people are to roadways. In addition, a limited number of monitoring stations might not accurately capture the true average ambient concentration, either because there are not enough monitoring stations or because they are not well situated throughout the air basin.

The close agreement in the calculated intake fractions for benzene and CO indicates that certain components of the underlying data are likely to be reasonably accurate. For example, if the emissions data I am using were significantly biased, it is possible but unlikely that the data for both benzene and CO would be biased the same amount. In addition, the agreement between the results presented in this work and in other works provides general support for the broad accuracy of the results reported here. However, the close agreement in the benzene and CO results does not intrinsically lend support to calculations that are applied to both chemicals. For example, if the population size I use is incorrect, it will have the same effect on both the benzene and the CO results.

## Conclusion

The intake fraction (iF) approach to exposure assessment characterizes the fraction of a pollutant's emissions taken in by people. The iF presented in this paper is for population exposure to atmospheric emissions from motor vehicle emissions in the South Coast Air Basin (SoCAB). However, the iF approach can be applied to individuals or subpopulations, and it can involve a multi-pathway, multi-media exposure assessment.

I calculated an iF of 48 per million, meaning that 0.0048% of SoCAB nonreactive emissions for motor vehicles are inhaled. The results for CO and benzene are similar (within 5%) and consistent with previous intake fraction studies, thereby lending support to the intake fraction approach. I have demonstrated the applicability of the approach to nonreactive gas emissions from a distributed source in the South Coast Air Basin by using the iF values for benzene and CO to estimate population intake of several nonreactive compounds found in motor vehicle exhaust.

There are several advantages to presenting exposure analyses in terms of the iF. The iF is a tangible concept that increases understanding and improves intuition about exposure assessments. It is a useful diagnostic tool to help validate empirical and modeled exposure assessments, and it can be used to summarize the importance of various transport mechanisms for each chemical being studied. Finally, the iF is an excellent tool for technical and nontechnical researchers to understand exposure assessment results. Policy makers want to make progress towards efficiently and effectively reducing human exposure to hazardous air pollutants, and the intake fraction is a valuable exposure metric for understanding how reductions in emissions relate to reductions in intakes.

Table 1: Summary of Ambient Concentration Data

	Carbon monoxide	Benzene
Number of data points	623,534	518
Percent non-detects	5%	6%
Precision	0.1 ppm	0.1 ppb
Detection limit	0.1 ppm	0.2 - 0.5 ppb
Average value	1.20 ppm	1.29 ppb

**Table 2 – Intake of Various Motor Vehicle Emissions**

		1,3-butadiene	acetaldehyde	benzene	formaldehyde	styrene	acrolein
SoCAB Emissions from motor vehicles	$t\ y^{-1}$	1067	1235	5482	3963	291	8
Lifetime	h	6	39	490	12	23	17
Reaction rate constant (k)	$d^{-1}$	4.1	0.6	0.05	2.1	1.0	1.4
Reactivity correction	-	28 - 47 %	71 - 85 %	97 - 99 %	43 - 63 %	60 - 78 %	52 - 72 %
iF for this pollutant	per million	13 - 22	34 - 41	46 - 47	20 - 30	28 - 37	25 - 34
Population intake	$kg\ y^{-1}$	14 - 24	42 - 50	253 - 257	80 - 119	8 - 11	0.2 - 0.3

**Notes:**

- 1) These results are for inhalation of primary motor vehicle emissions in the South Coast Air Basin. Degradation of primary emissions is included, but secondary formation is not.
- 2) Motor vehicle emissions in SoCAB are 42% of California motor vehicle emissions.
- 3) The reaction rate constant (k) is the reciprocal of the lifetime.
- 4) The reactivity correction, which is derived in Appendix 1, is multiplied by the intake fraction for a conserved pollutant to account for the presence of removal mechanisms. The intake fraction for a conserved pollutant is taken as 47.5 per million, which is the average of the results for benzene and carbon monoxide presented in the report. The range of values for the reactivity correction in this table are based on high and low values for the residence time of air in the air basin. The residence time of air in SoCAB is estimated in Appendix 1 as between 7 and 16 hours.

**Data Sources:**

California motor vehicle emissions	California Toxics Emission Inventory for 1996. See <a href="http://www.arb.ca.gov/toxics/cti/cti.htm">http://www.arb.ca.gov/toxics/cti/cti.htm</a> . Note that the data have been converted from tons per day to tonnes per year.
Atmospheric lifetime for 1,3-butadiene, acetaldehyde, benzene, and formaldehyde	EPA 1993 Motor Vehicle-Related Air Toxics Study. See <a href="http://www.epa.gov/otaq/toxics.htm">http://www.epa.gov/otaq/toxics.htm</a> .
Atmospheric lifetime for styrene and acrolein	CARB Toxic Air Contaminant Fact Sheets. See <a href="http://www.arb.ca.gov/toxics/tac/toctbl.htm">http://www.arb.ca.gov/toxics/tac/toctbl.htm</a> .
Percent of California emissions occurring in SoCAB	BC Singer and RA Harley. A fuel-based inventory of motor vehicle exhaust emissions in the Los Angeles area during summer 1997. Atmospheric Environment v34. 2000. 1783-1795.



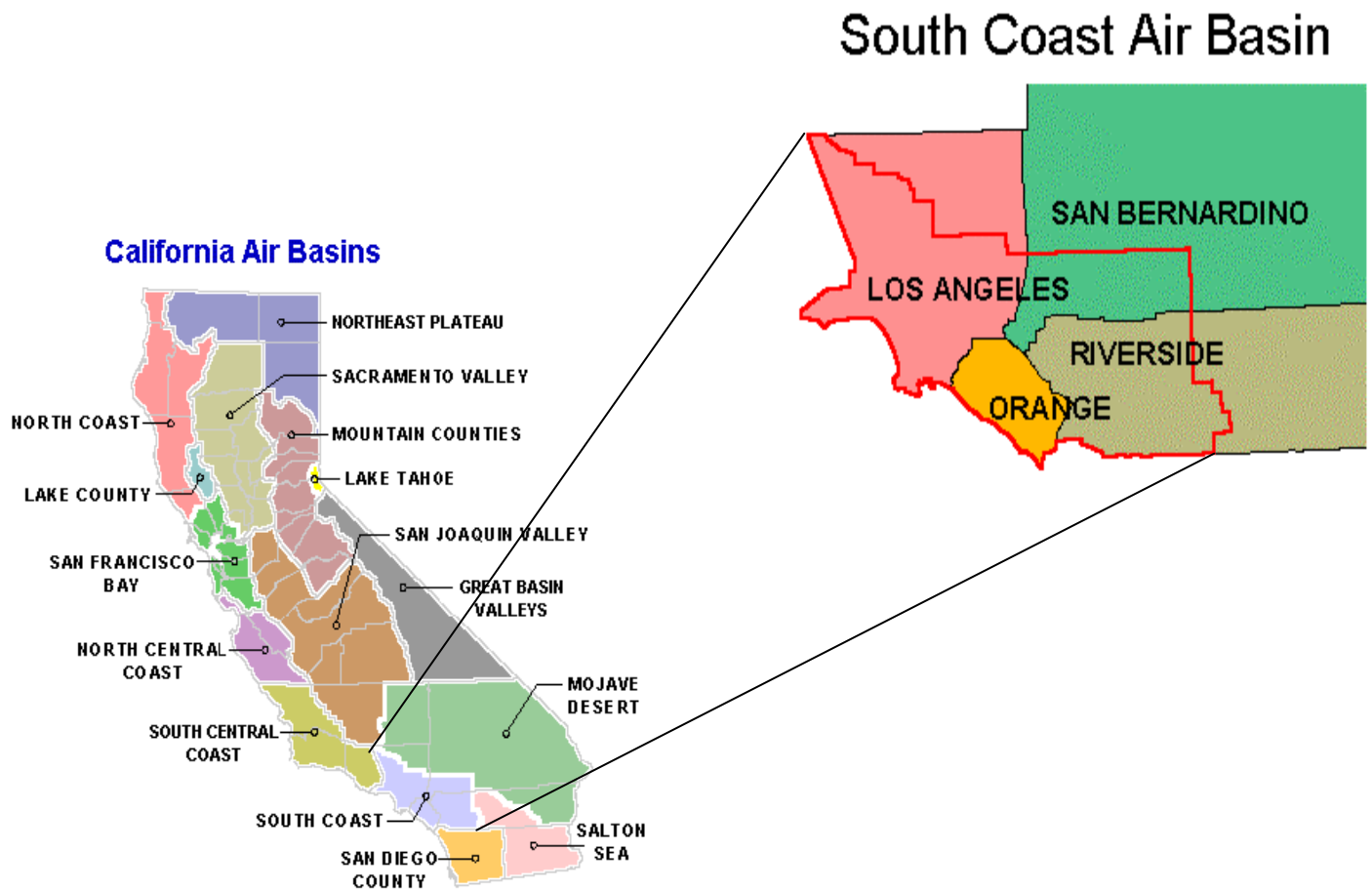
Table 3: Simplified Intake Fraction Analysis

		Carbon Monoxide	Benzene
Concentration	ppm	1.20	0.00129
Concentration	$\mu\text{g m}^{-3}$	1410	4.22
Ambient Concentration Attributable to Motor Vehicles	-	80%	70%
Breathing Rate	$\text{m}^3 \text{d}^{-1}$	12.2	12.2
Population	people	1.5E+07	1.5E+07
Intake	$\text{g month}^{-1}$	6.3E+06	1.6E+04
Emissions	$\text{g month}^{-1}$	2.0E+11	5.0E+08
Intake Fraction	per million	32	33

Table 4: Sensitivity Analysis

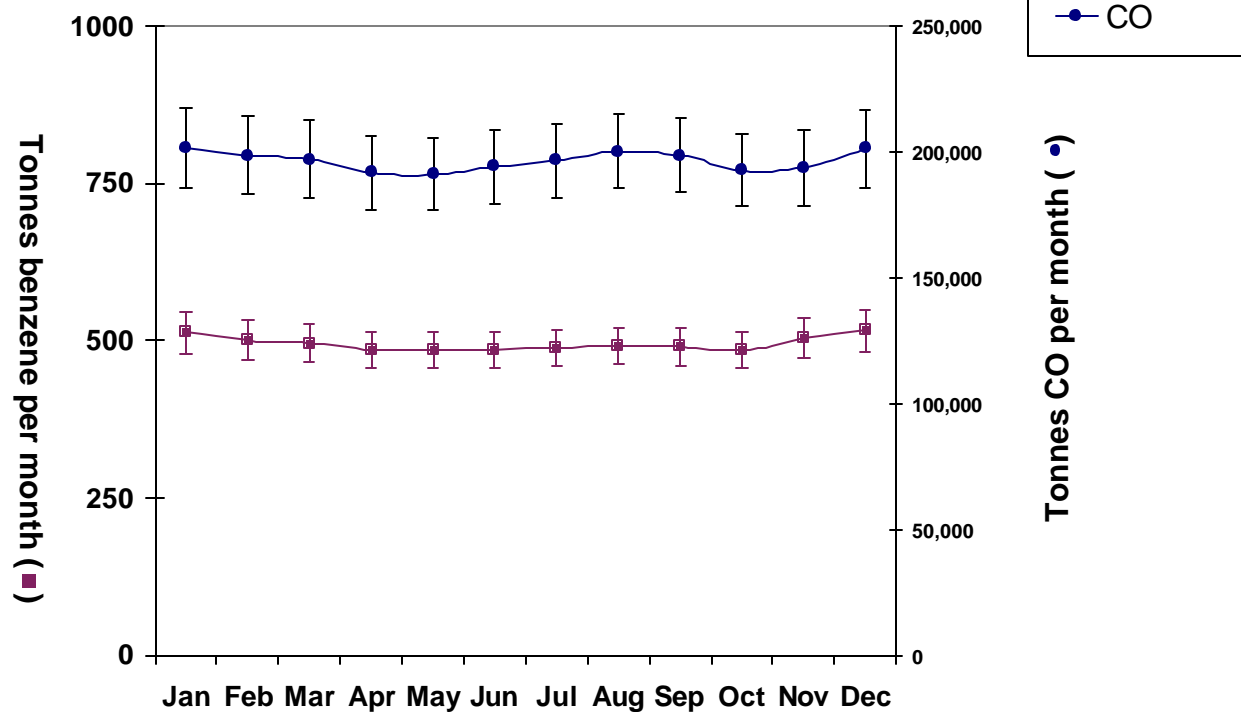
Parameter	Change in Parameter	Change in Intake Fraction
Concentration in vehicles	+ 20%	+ 6%
	- 20%	- 6%
	+ 100%	+ 30%
	-100%	- 30%
Time spent in vehicles	+ 20%	+ 6%
	- 20%	- 6%
	+ 100%	+ 30%
	-100%	- 30%
Concentration in residences with an attached garage	+ 20%	+ 8%
	- 20%	- 8%
	+ 100%	+ 40%
	-100%	- 40%
Population density at each specific monitoring station	+ 100%	-2% to + 8%
	- 100%	- 10% to + 2%

**Figure 1: Map of the South Coast Air Basin**

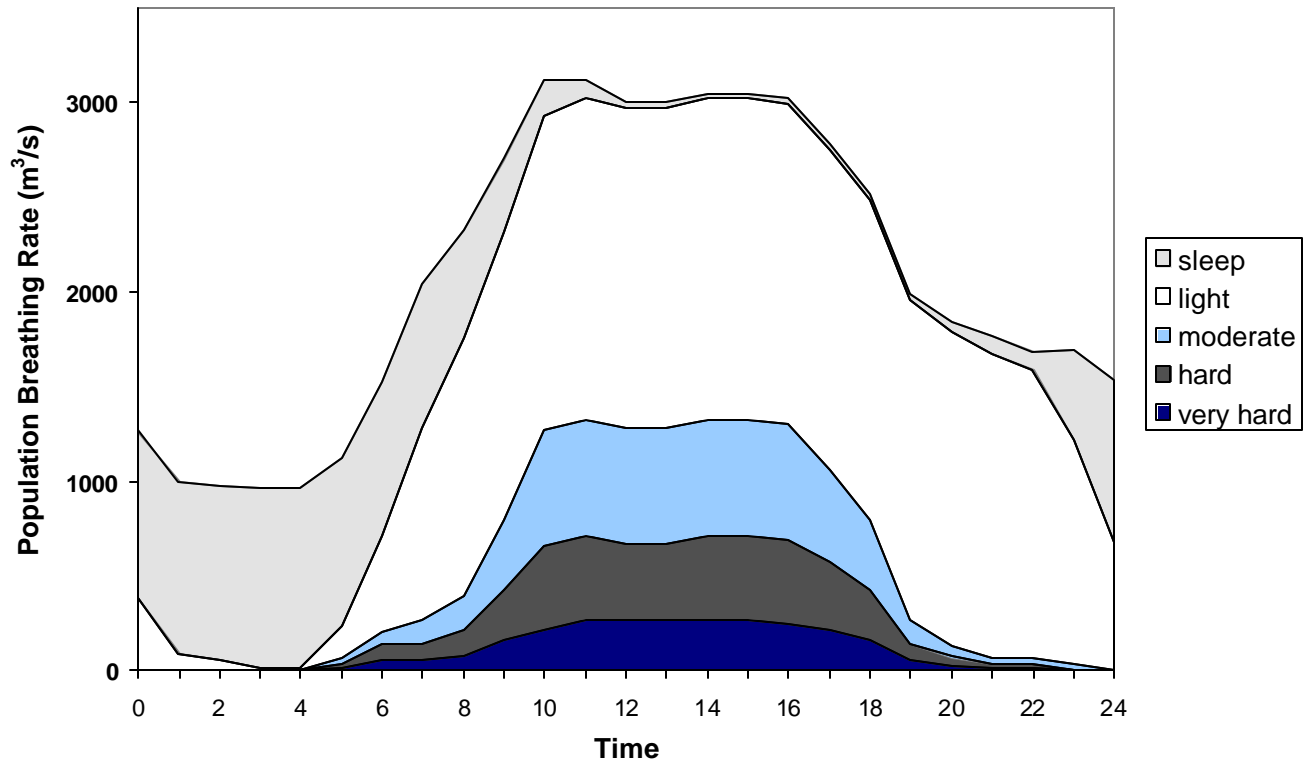


A map showing California's 15 air basins, and the South Coast Air Basin overlaid on the counties it includes. From <http://www.arb.ca.gov/emisinv/maps/statemap/abmap.htm>.

**Figure 2 - Motor Vehicle Emissions  
in the SoCAB 1996 - 1999**

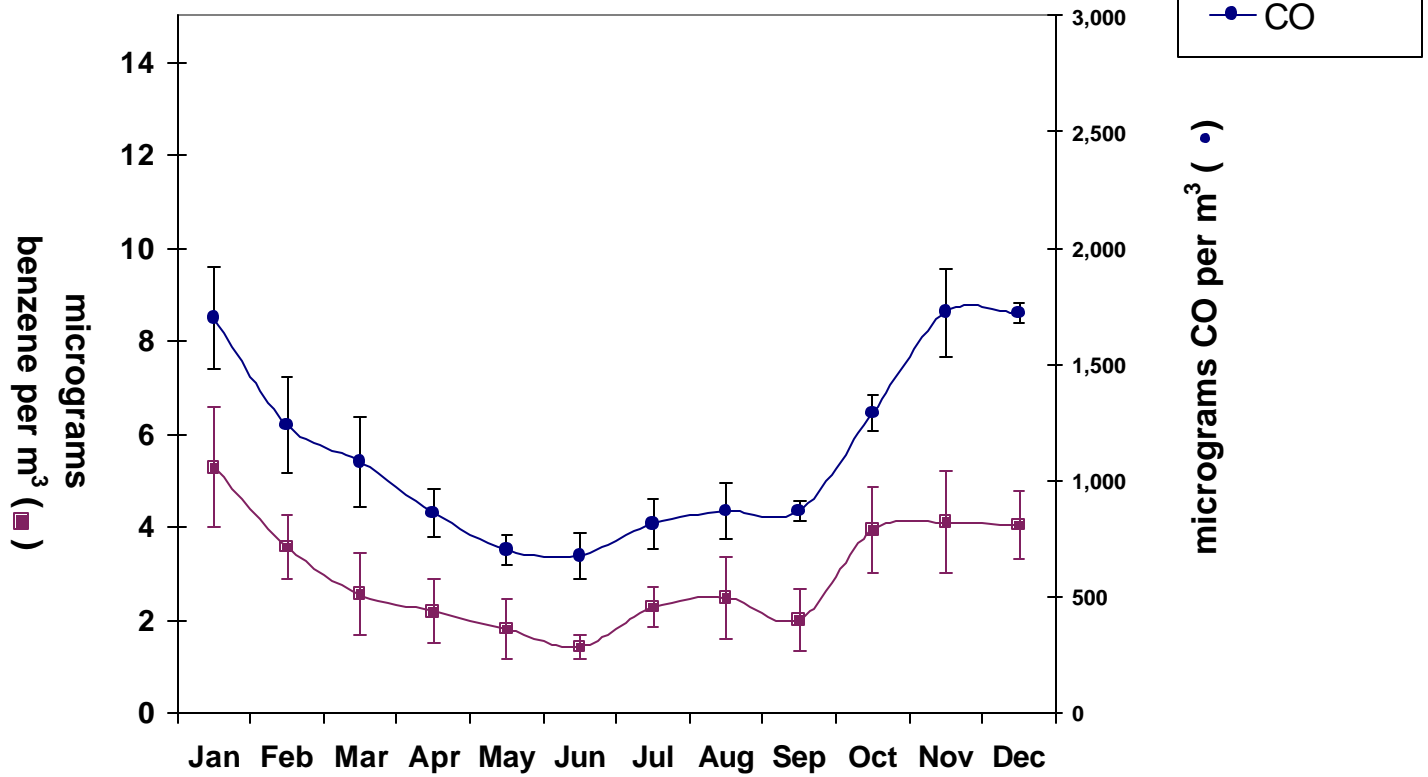


**Figure 3 - Hourly Breathing Rate  
by Time of Day and Activity Intensity**



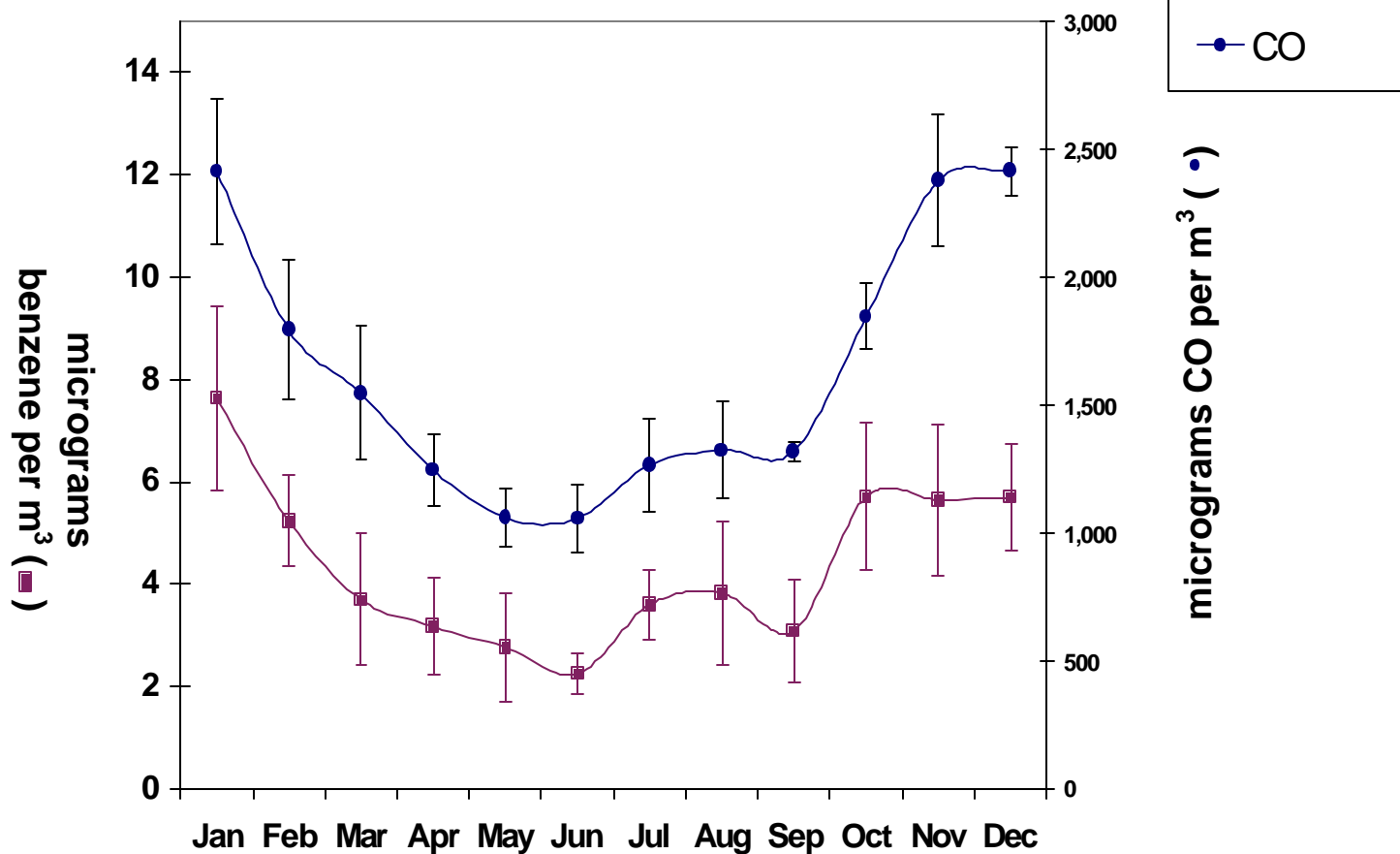
Layton (1993) gives breathing rates for five activity levels (sleep, light, moderate, hard, very hard) and the total number of hours spent in each of those activities. Figure 3 shows how these rates and hours were divided into the hours of the day.

**Figure 4a - Ambient Concentration  
Attributable to Motor Vehicles  
in the SoCAB 1996 - 1999**



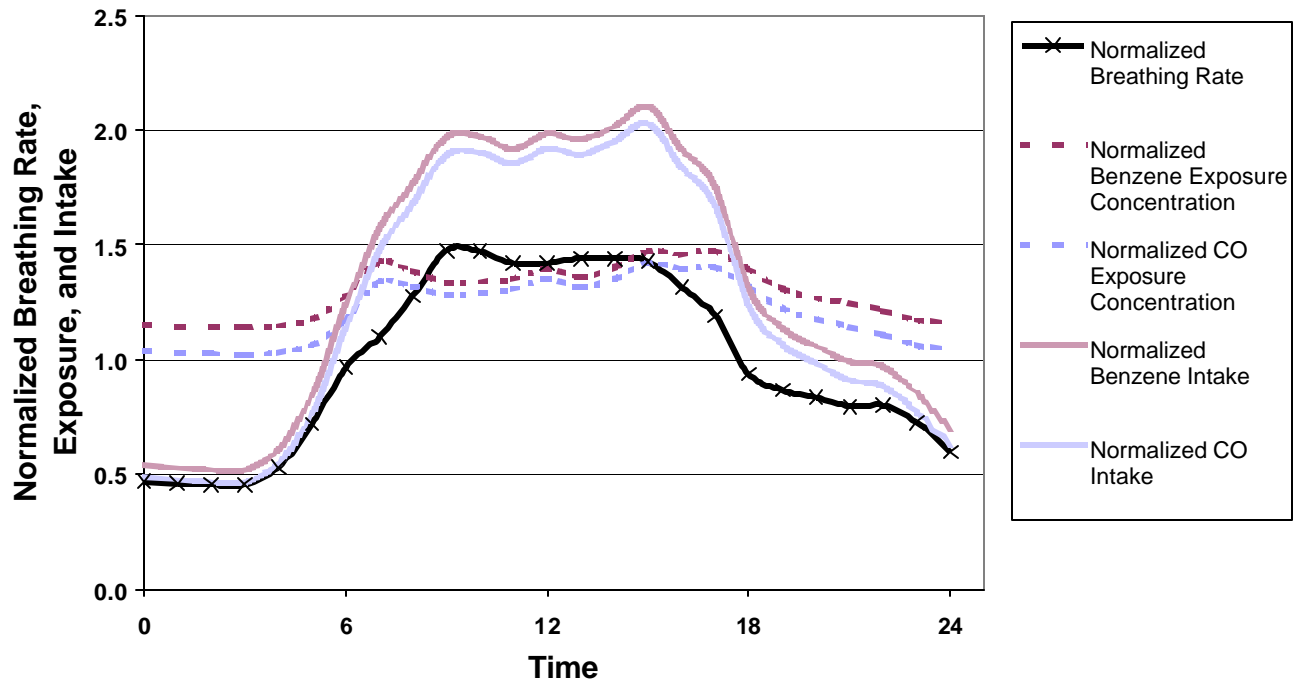
The ambient concentration attributable to motor vehicles, which is based on ambient air monitoring station data, shows a “U-shaped” profile due to the predominant meteorology. Summer conditions tend to disperse primary pollutants more efficiently than winter conditions.

**Figure 4b - Exposure Concentration  
Attributable to Motor Vehicles  
in the SoCAB 1996 - 1999**



Exposure concentration to benzene and CO attributable to motor vehicles shows the same pattern as the ambient concentrations in Figure 4b.

**Figure 5 - Hourly Variability in Breathing Rate, Exposure, and Intake**

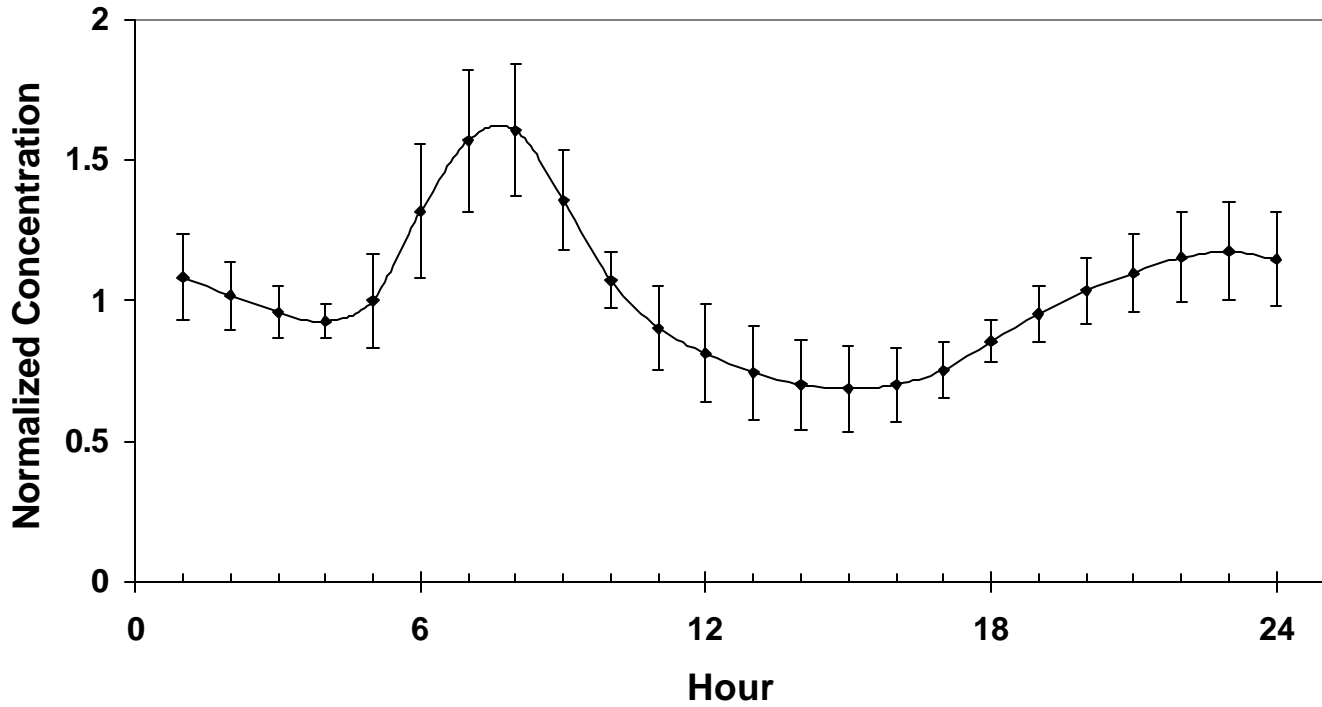


Normalized breathing rate is the hourly breathing rate divided by the average breathing rate. Normalized exposure concentration is the exposure concentration divided by the ambient concentration. Normalized intake, which is the product of the normalized breathing rate and the normalized exposure concentration, indicates the increase in the true intake, as compared to the simplified analysis, due to the combined influence of microenvironments and breathing rates.

This plot shows that the exposure concentration is greater than the ambient concentration at all times. Normalized benzene and CO intake is greater than one during the daytime, indicating that during the daytime, population intake is greater than ambient concentration times the average breathing rate. At night, population intake is less than ambient concentration times average breathing rate.

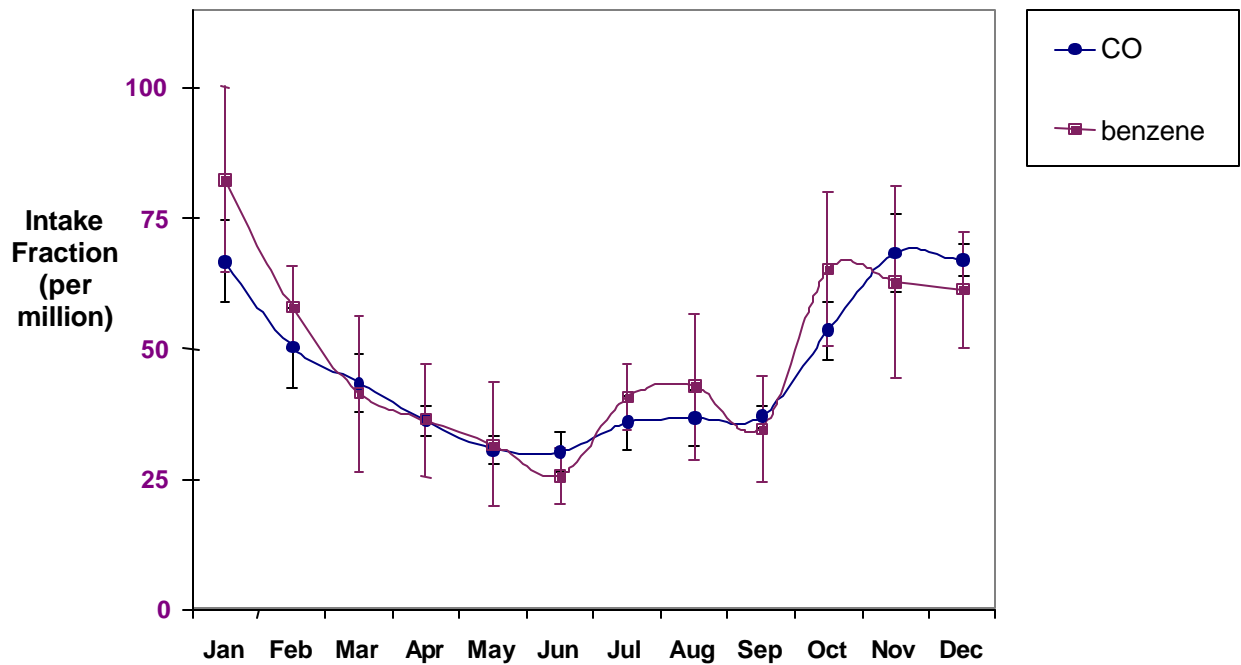


**Figure 6 - Typical Daily CO  
Concentration Profile**



Normalized concentration is the concentration in each hour divided by the average concentration. Concentrations are highest during the morning commute, when emissions are high and dispersion is low.

**Figure 7 - Intake Fraction for Motor Vehicles  
in the SoCAB 1996 - 1999**



Intake fractions for benzene and CO show the same pattern seen in Figures 4a and 4b. Consistency between the values for benzene and CO lends support to the intake fraction concept.

## Appendix 1: Box Model for Estimating Intake Fraction

Here I use a box model to estimate the intake fraction for a conserved and for a reactive compound. This approach indicates the influence of various factors on the calculated intake fraction. In a box model, the air in the basin is assumed to be well mixed and pollutant concentrations are assumed to be at steady state.

The population intake is given in Equation A1, and the mass balance equation for the box model is given in Equation A2. These two equations are combined in terms of the intake fraction in equation A3.

$$\text{Total Intake rate} = CQ_B P \quad \text{A1}$$

$$\begin{aligned} \text{Emission rate} &= \{\text{Loss rate due to reaction}\} + \{\text{Removal rate by advection}\} \\ E &= \{kCV\} + \{QC\} \end{aligned} \quad \text{A2}$$

$$\text{Intake Fraction} = \frac{\text{Total Intake Rate}}{\text{Total Emissions Rate}} = \frac{CQ_B P}{E} = \frac{Q_B P}{kV + Q} = \frac{Q_B \rho t}{H(k t + 1)}$$

A3

Variables:

$C$  = Concentration increment owing to emission source ( $\text{g}/\text{m}^3$ );

$E$  = Emission rate ( $\text{g}/\text{s}$ );

$u$  = Wind speed ( $\text{m}/\text{s}$ );

$L$ ,  $W$ , and  $H$  = Length, width, and height of the air basin (meters);

$A$  = Area of the air basin ( $\text{m}^2$ ) =  $LW$ ;

$V$  = Volume of the basin ( $\text{m}^3$ ) =  $LWH$ ;

$Q$  = Flux of air through the basin ( $\text{m}^3/\text{s}$ ) =  $WHu$ ;

$\tau$  = Residence time of air in the basin ( $\text{s}$ ) =  $V/Q = L/u$ ;

$k$  = First-order reaction rate constant ( $\text{s}$ ), where  $k=0$  for a conserved pollutant;

$P$  = Population exposed in air basin (people);

$\rho$  = Average population density ( $\text{people m}^{-2}$ ) =  $P/A$ ; and

$Q_B$  = Average breathing rate per person ( $\text{m}^3 \text{s}^{-1}$ ).

Note that for a relatively nonreactive compound,  $k\tau \ll 1$ . In this case, equation A3 reduces to  $iF \sim Q_B \rho \tau / H$ . For a highly reactive compound,  $k\tau \gg 1$ . In this case, equation A3 reduces to  $iF \sim (Q_B \rho) / (Hk)$ . A compound is defined as being moderately reactive when  $\tau \sim (1/k)$ , i.e., when the reciprocal of the reaction rate constant is similar in magnitude to the residence time. In equation A3, the term  $(k\tau + 1)^{-1}$  stands out as a “reactivity correction term.” If the intake fraction is known for a nonreactive compound, multiplying by this term will yield an intake fraction estimate that accounts for the reactivity of a specific compound.

A3 can be rewritten as

$$\text{Intake Fraction} = \frac{Q_B r}{H} T$$

A4

where  $T$  is the characteristic time for the pollutant to be removed from the system. For a nonreactive compound, the characteristic time is  $T = \tau$ . For a highly reactive compound, the characteristic time is  $T = k^{-1}$ . For a moderately reactive compound, the characteristic time is given by  $T^{-1} = k + \tau^{-1}$ .

Table A1 first uses  $iF = (Q_B P) / (uHW)$  to calculate the intake fraction for a conserved pollutant, and then applies the “reactivity correction term”  $(k\tau + 1)^{-1}$  to predict the intake fraction for a reactive pollutant with  $k^{-1} = 80$  hours. This reactivity term varies from 18% to 92%, meaning that the intake fraction for the reactive pollutant is 18 – 92% of the intake fraction for a conserved pollutant.

The first entry for the SoCAB columns represents the product of the wind speed and mixing height. Unfortunately, mixing height data are not available for the South Coast Air Basin. The values used in Table A1 ( $195 - 1,300 \text{ m}^2 \text{ s}^{-1}$ ) are the 5% and 95% values from the distribution of harmonic means for the 75 cities in the EPA's SCRAM database of mixing heights (EPA). The mixing heights for the downwind regional intake (1,200 – 12,000 m) represent mixing occurring throughout 10 – 100% of the troposphere, and the mixing height for the US intake (12,000 m) assumes mixing throughout the troposphere. The wind speeds ( $2.36 - 5.50 \text{ m s}^{-1}$ ) represent the 5% and 95% distribution of harmonic means for the 75 cities in the EPA's SCRAM database of wind speeds. These wind speeds are the average over the mixing zone rather than being the surface wind speed. As is discussed in the text, the reaction rate ( $k^{-1} = 80$  hours) represents a compound that is more reactive than benzene in the summer.

The  $iF$  for a conserved pollutant in the SoCAB is estimated at 12 – 82 per million using the box model. This range includes both the results above of  $\sim 48$  per million for benzene and CO, and the results presented in Table 3 of  $\sim 33$  per million from the simplified analysis. Consistency between the box model and these two results lends support to the box model approach.

**Table A1: Intake Fractions Using a Box Model**

		SoCAB		Downwind Regional		USA	
H				<b>1,200</b>	<b>12,000</b>	<b>12,000</b>	<b>12,000</b>
u				<b>2.36</b>	<b>5.50</b>	<b>2.36</b>	<b>5.50</b>
Hu	$\text{m}^2 \text{s}^{-1}$	<b>195</b>	<b>1,300</b>	<b>2,832</b>	<b>66,000</b>	<b>28,320</b>	<b>66,000</b>
A	$\text{miles}^2$	6,729	6,729	32,340	32,340	3,537,000	3,537,000
A	$\text{m}^2$	<b>1.7E+10</b>	<b>1.7E+10</b>	<b>8.4E+10</b>	<b>8.4E+10</b>	<b>9.2E+12</b>	<b>9.2E+12</b>
W	m	<b>1.3E+05</b>	<b>1.3E+05</b>	<b>2.9E+05</b>	<b>2.9E+05</b>	<b>3.0E+06</b>	<b>3.0E+06</b>
P	people	<b>1.5E+07</b>	<b>1.5E+07</b>	<b>1.3E+06</b>	<b>1.3E+06</b>	<b>2.81E+08</b>	<b>2.81E+08</b>
$Q_B$	$\text{m}^3 \text{d}^{-1} \text{person}^{-1}$	12.2	12.2	12.2	12.2	12.2	12.2
$PQ_B$	$\text{m}^3 \text{s}^{-1}$	2.1E+03	2.1E+03	1.8E+02	1.8E+02	4.0E+04	4.0E+04
uHW	$\text{m}^3 \text{s}^{-1}$	2.6E+07	1.7E+08	8.2E+08	1.9E+10	8.6E+10	2.0E+11
iF	-	8.2E-05	1.2E-05	2.2E-07	9.6E-09	4.6E-07	2.0E-07
<b>iF</b>	<b>per million</b>	<b>82</b>	<b>12</b>	<b>0.22</b>	<b>0.010</b>	<b>0.46</b>	<b>0.20</b>

L	m	1.3E+05	1.3E+05	2.9E+05	2.9E+05	3.0E+06	3.0E+06
$k^{-1}$	h	80	80	80	80	80	80
k	$\text{h}^{-1}$	0.0125	0.0125	0.0125	0.0125	0.0125	0.0125
u	$\text{m s}^{-1}$	2.36	5.50	2.36	5.50	2.36	5.50
$\tau$	h	16	7	34	15	356	153
$(k\tau+1)^{-1}$	-	84%	92%	70%	85%	18%	34%
<b>iF</b>	<b>per million</b>	<b>69</b>	<b>11</b>	<b>0.16</b>	<b>0.008</b>	<b>0.08</b>	<b>0.07</b>

## Appendix 2: Intake Fraction by Month

Table A2 summarizes the data used to calculate the intake fraction in each month. The data for benzene and carbon monoxide are shown separately. For each chemical, the intake fraction is calculated first based on the ambient concentration, and then based on the near-source contributions. These two parts are then added to determine the total intake fraction.

Note that there are several analyses that are included in the near-source intake fraction, beyond the issue of near-source exposures. The name “near-source” is used here to indicate that there are many additional analyses included, rather than just using the ambient concentrations. These additional analyses include:

- using the hourly CO concentration rather than the monthly average;
- applying the typical hourly CO concentration profile in each month and year to the 24-hour benzene concentration;
- using hourly breathing rates; and
- accounting for microenvironment concentrations.

Table A2 provides the data used in this analysis, the ambient concentration intake fraction, the near-source intake fraction, and the sum of these intake fractions.











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